40792/2

UNIVERSITY AT BUFFALO
STATE UNIVERSITY OF NEW YORK

Department of Electrical and Computer Engineering
School of Engineering and Applied Sciences
312 Bonner Hall
Box 601900
Buffalo, New York 14260-1900
(716) 645-2422 ext 1202

sent 1996 February
due 1996 February 15
R&T Number: s400024srd
ONR Grant No:N00014-93-1-0483
Grant Period 15 Feb 93 - 19 June 96
"High Power Electronics Institute"
RF of SUNY 150-4758A (ONR2c)

THIRD ANNUAL REPORT (Attached)

Effort Period: 15 Feb 1995 - 15 Feb 1996

TO: (3 copies): Scientific Officer Code: 1132P
Gabriel D. Roy
Office of Naval Research
800 North Quincy Street
Arlington, VA 22217-5000

Additional Distribution:
(1 copy): Administrative Grants Officer
Office of Naval Research
Resident Representative N62927
Administrative Contracting Officer
33 Third Avenue - Lower Level
New York, NY 10003-9998

: (1 copy): Director, Naval Research Laboratory
Attn: Code 2627
Washington, DC 20375

: (2 copies): Defense Technical Information Center
Building 5, Cameron Station
Alexandria, VA 22304-6145

FROM: Rich Dollinger
Acting Deputy Director

Richard E. Dollinger

COPY: Jim Sarjeant, Principal Investigator
: George Lee, Principal Investigator
: Javaid Laghari, Investigator
: Ann Salter, Sponsored Programs
: Travel

ONR2c,"AY3cover",005 - Cover Letter -

THIRD ANNUAL REPORT

Effort Period: 15 Feb 1995 - 15 Feb 1996

R&T Number: s400024srd Grant Period: 15 Feb 1993 - 19 June 1996

ONR Grant No.: N00014-93-1-0483 Princ. Inv.: Sarjeant and Lee

a. Description of scientific research goals

Develop a physical understanding of the nature of the processes responsible for multifactor stress life aging of semiconductor materials and their oxide insulators in support of the BMDO/ONR High Power Electronics Program.

b. Significant results in the past fiscal year

The initial proof of concept and development of a new and novel method to study the aging of semiconductor materials and their oxide interfaces under dc through high frequency fields (up to 100 kilohertz ac) has been successful. This new balanced bridge technique approach yields up to 100 times enhanced sensitivity in comparison to all other aging metholodgies, and, at the same time permits direct correlation with dc and low frequency ac aging by virtue of the tranportable test fixturing. This year also saw successful accelerated dc aging, relevant to the next generation of dc high power solid state radars and power conditioning systems.

The initial ac aging experiments to investigate the effect of continuous high frequency electrical stress upon insulating materials and their related insulating oxide coating/interfaces was completed. The data provided preliminary complex aging rates and mechanism identification, as well as guidance as to how to proceed with high frequency ac insulation aging in a methodical manner. It was also found that corona voltages exhibited a reduction with increasing frequency of the applied stress, due to acceleration of the ionization process at the interface and within the material substructure.

Concern for the reliability of high power electronics components/systems demanded the investigation of the insulator component's aging and failure modes under service conditions which imposes a multitude of deterministic stress factors such as thermal and electrical stresses. Thermal stresses, whether external or internal (localized heating), are dominant in high power electronics systems and the investigation of their effects within service temperature ranges is integral to the understanding and modeling of the performance and reliability of the high power electronics system as a whole.

The investigation of the effects of thermal aging on the properties of insulating materialsincluing surface and bulk diagnosis, determination of properties and statistical analysis of the dc failure times to infer and model their functional relationship to the sequentially applied stresses, was thus carried out. The material selected in this study was recommended by the US industry, and is an insulating film presently utilized in numerous high power electronics applications, namely capacitor grade biaxially-oriented isotactic polypropylene film in the 25 μm (1 mil) range.

Surface analysis utilized state of the art diagnostics such as Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM) for microscale topographic mapping and Electron Scanning for Chemical Analysis (ESCA) to detect the presence of any deposited elements which may alter the chemical surface characteristics. The ESCA spectra of surface elements (up to 50 A depth) of thermally aged and unaged film samples confirmed the oxidation of the film surface as a result of aging as shown in Figure 1. Samples aged for up to 100 hours at either 90 °C or 110 °C did not show any trace of surface oxygen. Samples aged at 90 °C for 300 and 500 hours showed an increase in the concentration of surface oxygen (1.12 and 1.24%, respectively), and for samples aged at 110 °C higher percentages (1.6 and 1.7%, respectively), were detected. Although the oxygen percentages are small, nonetheless, the slightest oxidation of the uppermost surface layer will have profound effects on the interface properties transforming it from a low energy to a high energy, and consequently affecting the interaction mechanisms and characteristics with other components of the test system. AFM images indicated altered surface roughness measures for the aged samples, which were subsequently supported by results of the SEM images.

The mean dc breakdown voltages correlated with the surface oxygen trend detected by ESCA. This is shown in Figure 2. Generally, the breakdown voltages increased with increasing aging time and temperature. However, a t-test did not conclude a significant difference between the breakdown voltages for the control and the aged film samples for 100 hours at either 90 °C or 110 °C. It is, therefore, reasonable to conclude that the change of surface properties, in the presence of the electric field, leads to interfacial space charge effects. For example, the formation of negative space charge at the electrode-film interface decreases the effective field increasing the apparent breakdown field.

SEM images of the breakdown region in control and aged film samples strongly suggested the existence of two distinct breakdown/failure mechanisms for the control and aged film samples. A seemingly bulk-limited mechanical brakdown was deduced for the control samples, while localized interfacial breakdown was deduced for the thermally aged samples due to the altered surface properties of the thermal aged film samples and consequent space charge effects in the presence of the applied electrical stress field.

The statistical analysis of the lifetimes (time to failure) included graphical and analytical methods for assessment of candidate lifetime distributions: Weibull and Lognormal; Analysis of Variance (ANOVA) to subdivide variation in the replicate lifetime data into systematic variation due to significant stress variables (thermal aging variables and dc stress) and that due to

error/noise as a result of inherent stochastic spatial inhomogeneities. Finally, modeling of the functional relationship between the obtained dc lifetimes and functions of the sequentially applied stresses was attempted using phenomenological semiempirical laws available in the insulation literature. The results indicated that a significant 82% of the variation in the data was due to the deterministic stress variables. An exhaustive search among possible transformed-linear and nonlinear models found a multiplicative effects linear model to predict the dc lifetimes with sufficient accuracy based on maximum likelihood analysis. Figure 3 shows the actual versus the predicted life lines for the test conditions considered.

The present investigation provided a deep understanding of the effects of thermo-oxidative aging within service temperature range (up to 110 °C) of the high power electronics systems on the properties and characteristics of the insulating material. Generally, the effects were desirable as the lifetimes and breakdown voltages increased or at worst remained unchanged. The chosen insulating material proved very resiliant to such stresses making it a safe choice for many high power electronics applications within the limitations of the test conditions. The study also revealed the complexity of modeling the functional relationship between failure times and stress factors as many determined stochastic variables convolve leading to failure

c. Plans for next year's research

The comprehensive aging methodology being developed for semiconductor systems and their related insulating oxide coatings/interfaces, initiated this year, will be continued. The new emphasis will be upon materials of relevance to high power solid state electronics, interfaces, connectors, and power distribution and conditioning systems for next generation BMDO/ONR systems. Actual solid state topologies will be used to anchor the transition of the aging methology to real world systems, in collaboration with other researcher's active under BMDO support - both industrial and academic. Of particular interest is the aging under simultaneous DC+AC conditions, representative of a large number of emerging BMDO applications.

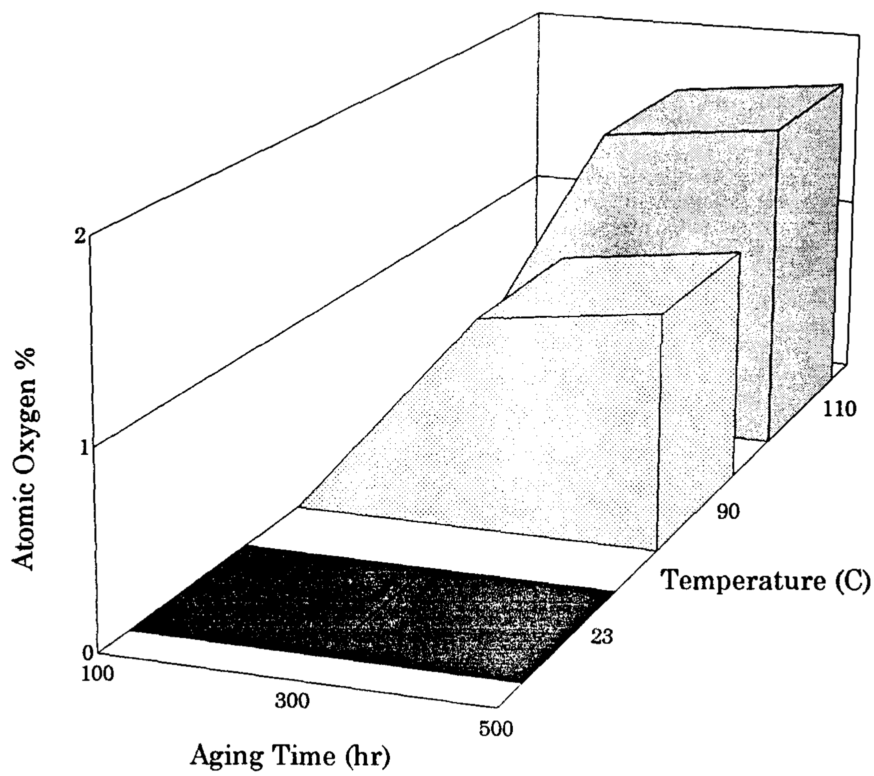
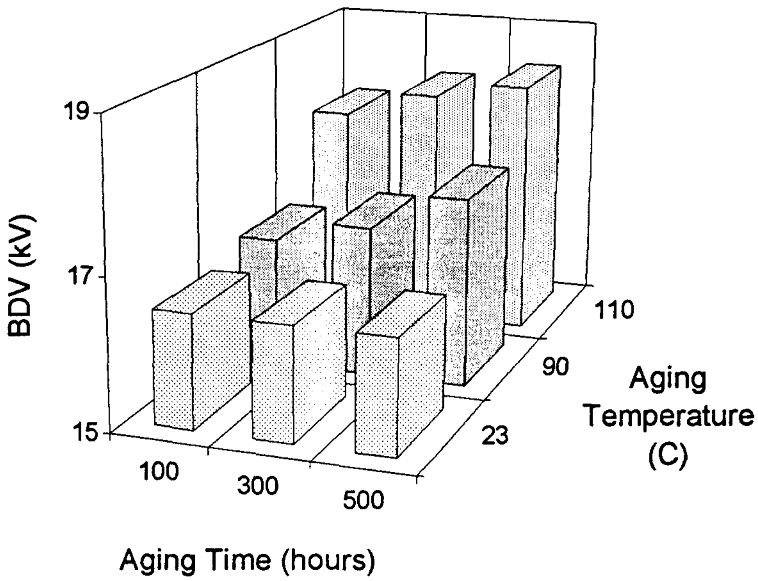


Figure 1. Oxygen percentages on sample PP film surfaces as detected by ESCA.



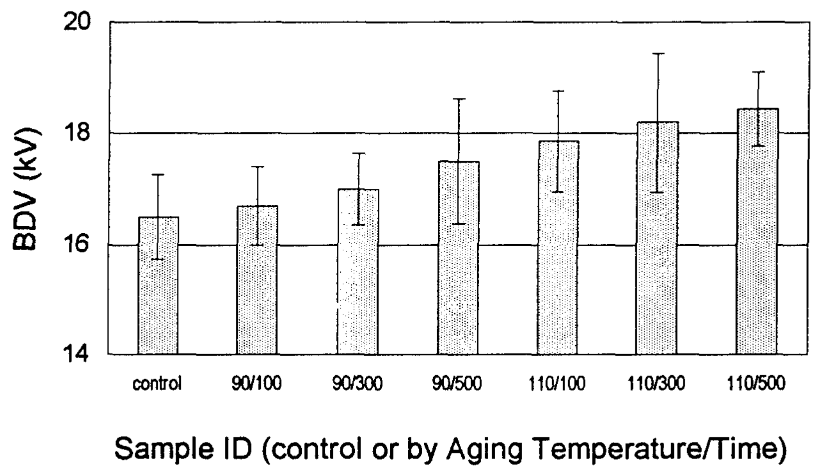
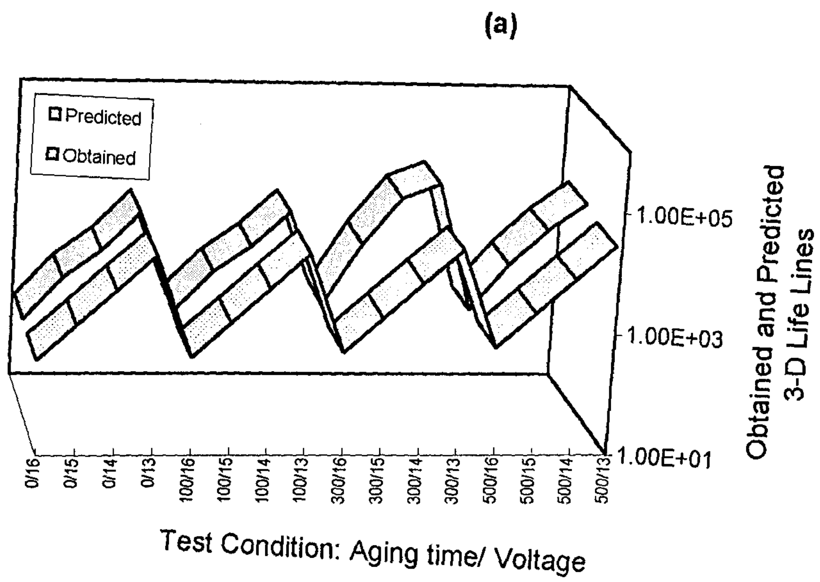


Figure 2. Mean dc breakdown voltages for tested PP film samples at shown test conditions.



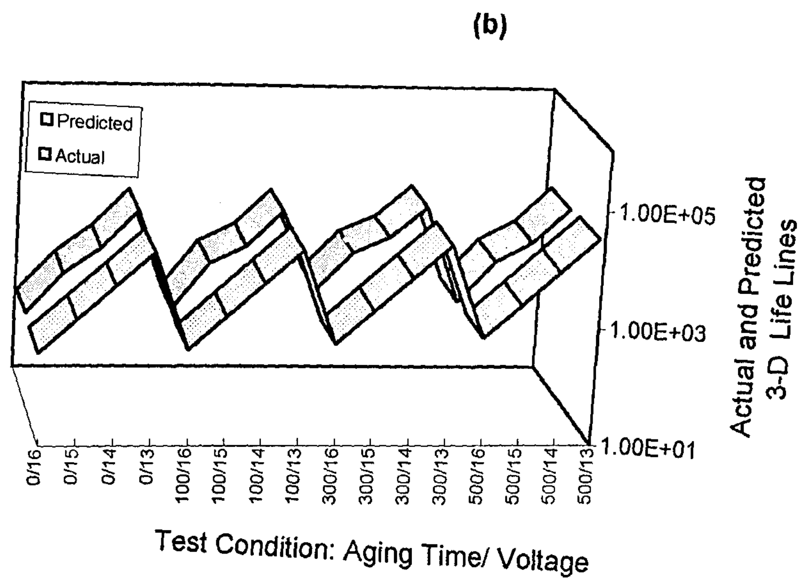


Figure 3. Predicted and obtained 3-D life lines for thermally aged film samples at 90 °C (a) and 110 °C (b). Control values are shown at zero aging time for reference.

LIST OF REFEREED PUBLICATIONS

Attachments:

1. W. J. Sarjeant, "Capacitors: Aging and Failure - A Review," CSC'2 Proceedings, 2nd International Conference on Space Charge in Solid Dielectrics, Antibes-Juan-Les-Pins, 2-7 April 1995, ASPROM IEEE SfV, pp. 84-92.

2. Amany N. Stokes, "An Investigation of Aging Mechanisms and Statistical Life Models for Polypropylene Films," State University of New York at Buffalo, Ph.D. Dissertation, June 1995.

DEGREES GRANTED

1.	A. Stokes,	June 1995	Ph.D.
2.	J. Chau,	March 1995	Master-of-Engineering

Supplément à la Revue "Le Vide : science, technique et applications" - N° 275, Mars 1995

CAPACITORS : AGING AND FAILURE - A REVIEW

W. Sarjeant
Electrical and Computer Engnieering, State University of New York at Buffalo - Bonner Hall, Room 312 - Buffalo/NY 14260-1900 (USA) - Tel. (716) 645-2422 ext. 1208

ABSTRACT

The fundamentals of capacitor dielectric technology and the aging of laminate insulation structures that form elements of this technology are reviewed. The failure processes in highly stressed high energy density capacitors attributed to a number of mechanisms, Intrinsic, Electromechanical, *Streamer*, *Thermal*, *Space Charge* and *Partial* Discharge breakdown are discussed. State of the art diagnostic measurement techniques available and those developed at the Dielectrics Laboratory at the State University of New York at Buffalo to evaluate the influence of space charge and partial discharge injection at frequencies to 10kHz are presented.

INTRODUCTION

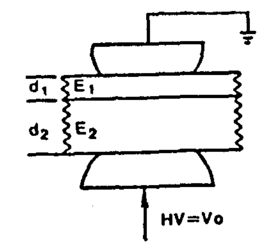
Capacitors allow electrical energy *to be stored over* a long charging time and then released over much shorter periods under controlled conditions. A typical construction of the capacitor is that of two conducting plates separated by layers (laminates) of dielectric material as shown in figure 1. The application of potential difference across plates leads to polarization of charge and thus energy storage. In the development of various high power electronics components, the area of prime interest is multihertz, long-life (high reliability), low inductance capacitors. Failure of electrical components used in various high power electronic systems is often attributed to the failure of the electrical insulation/dielectric due to the presence of degrading stresses such as electrical, thermal and environmental. The critical issue of system quality and reliability demands the understanding of failure and aging mechanisms by diagnosing, evaluating, and modelling appropriately selected properties of the electrical insulation polymer and its interaction with other components within the insulation system.

The complexity of modelling dielectric failure arises from the fact that there are many factors, known or unknown, at play. Figure 2 depicts such a black box system displaying the inputs comprised of controlled stress variables and a multitude of uncontrolled stochastic factors. The stochasticity of the every component and mechanism convolve to form the resulting output. The objective of the engineer is to model only the effect of the applied stress variables on the response variable of the output by assuming an *ideal dielectric* that is completely homogeneous. For practical purposes, such an assumption has limited application though it simplifies the problem significantly. It is clear that many assumptions about the system's random components are necessary to simplify analysis. Modelling the practical insulation system that is far from ideal while also bridging the gap to the purely theoretical treatments, utilizing available technology and statistical tools for conducting and analyzing cost and time effective experiments are all

CSC'2 Proceedings, 2nd International
Conference on Space Charge in Solid
Dielectrics, Antibes-Juan-Les-Pins,
2-7 April 1995
ASPROM IEEE SfV

Attachment #1 - Page 1 of 5

dimensions of the problem. Most approaches so far have been empirical while theoretical models struggle to account for the stochastic nature of the many factors that convolve to cause failure in practical systems [1].



$$E_1 = \frac{V_o}{d_1 + d_2(\frac{\epsilon_1}{\epsilon_2})}$$

$$E_2 = \frac{V_o}{d_1(\frac{\epsilon_2}{\epsilon_1}) + d_2}$$

Figure 1. A two-dielectric laminate system.

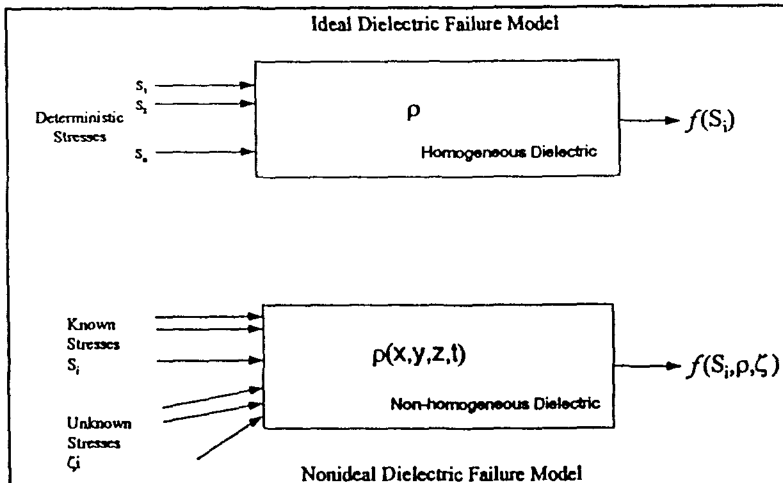


Figure 2. The complexity of modelling dielectric failure in a practical insulation system vs. the simplicity of the assumed ideal model by the engineer.

Approaches to modelling breakdown in the literature have been either via mathematical (semi-empirical) curve fitting of experimental and in-service cable data, or via theoretical studies of degradation mechanisms as summarized in Figure 3.

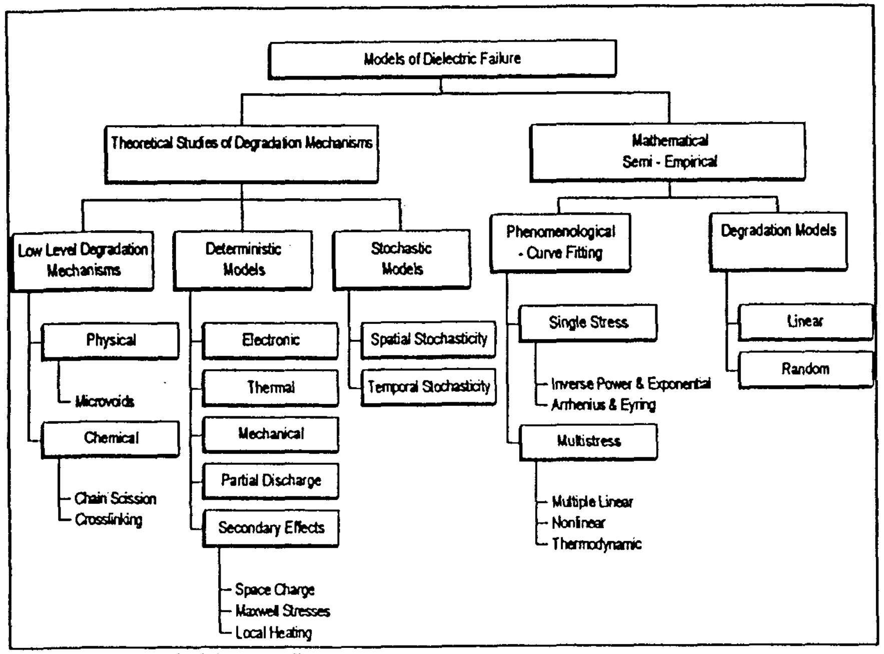


Figure 3. Models of Dielectric Failure

Space Charge is well known to affect and distort local high fields leading to dielectric and component failure and breakdown. The effects are seen in the case of constant stress DC fields as the time permits build up of charge that is either injected homocharge at electrode-insulator interface or results from deep trapped bulk charge and possibly void discharge. It is a function of the insulation material itself, the stress environment conditions such as temperature, impurities among many factors. The theory is rather well developed and reviews can be found in [2]. Recently [3], the development and utilization of several practical and noninvasive methods/techniques has allowed quantitative analysis of space charge via measurement of an induced current that is directly proportional to the space charge profile. Some of those methods include the LIPP (Laser Induced Pulse Pressure) method, PIPS (Piezoelectrically Induced Pressure Step method), the PEA (Pulsed Electroacoustic) method, the TP (Thermal Pulse) method, the TSP (Thermal Step Pulse) method, and the LIMM (Laser Intensity Modulation Method). This allowed further insight into the possible operating mechanisms leading to breakdown. Further research into that area remains needed to understand and model how space charge affects pre-breakdown processes leading to dielectric failure. The work done by [4] is an example.

All discharges are a result of ionization processes as depicted in figure 4, initiated by a few original free char carriers inherently present in the dielectric. Corona and partial discharges are well known aging drivers in A AC+DC, and low voltage pulse transformers while space charge dominates in DC applications.

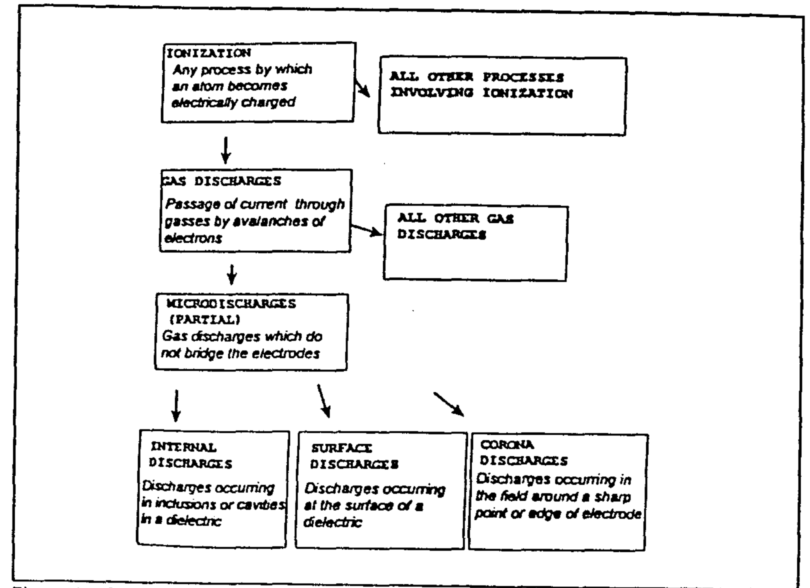


Figure 4. Ionization Processes and Charge Formation

Partial Discharges and Space Charge Injection

Charge carriers under the influence of a high electric field can be injected into the dielectric material from the surface of an electrode as well as due to PD inception. These carriers are transported through the dielectric, and are captured at the opposing electrode. Charge injection is not an impulse breakdown process, but is a continuous conduction phenomenon similar to intrinsic or extrinsic conduction except the origin of the carriers is entirely external to the dielectric, and the current is hyperlinear with the electric field. Two types of injection processes that have been specifically identified and described are Schottky and Tunnel injections [5]. Regardless of the physics at the electrode - dielectric interface, the result is a current density in the dielectric that can approach a space charge limited condition, where the electric field at the injection site is reduced to a small value by the proximity of space charge. Because charge injection is a field-controlled process, the current density will be maximum at the high field sites such as the foil edges of a foil wound capacitor. The conduction path follows the electric field vector and injected current flows from a foil edge to the opposing foils through the multiple layers of the dielectric. If the charge injection process can become established during the rise or fall times of voltage transients across the foils, the reduction of the electric field at the foil edge would increase the partial discharge inception voltage. Although

the time constants for the distribution of space charge during field-controlled injection are significantly shorter than the ϵ/σ (permittivity/conductivity) ratio modeling the capacitance as in figure 4, the values are large compared to the millisecond charge *times* and *nanosecond discharge times of pulse discharge capacitor service* [6].

Using the transport time for a slow electron under the influence of a 4-kv per mm field to travel a distance of 25 mm as a measure of the injected space charge formation time, the following equation is obtained,

$$t_f = \frac{d}{E \cdot \mu} \qquad (1)$$

where μ is the carrier mobility, d is the dielectric carrier thickness, E is the electric field near the foil edge (spatial average), and t_f is an estimate of space charge formation time. The mobility is known to be in the range of 10^{-12} - 10^{-16} $m^2 v^{-1} s^{-1}$ for most dielectrics and is relatively low because the carriers spend most of the time in the shallow traps so the mobility is an effective value. The range of t_f is then calculated to be $6 \times 10^{-7} < t_f < 6 \times 10^{-1}$ s.

The *hypothesis that space charge associated with field-controlled injection* reduces the electric field intensity at the high field sites on the foil edges and hence increases partial discharge inception voltage has been supported by laboratory experiments, sample results of which are summarized in figure 5. For details of the experiments, see [6].

It is hypothesized that when voltage is applied across the foils faster than space charge can form and reduce the field at the foil edge, partial discharges occur and create an impulsed, spatial distribution of charges in the regions near the foil edge, reducing the electric field below the partial discharge inception threshold. For conditions where the rate of voltage is negative (decreasing voltage), if the transient voltage is fast enough in fall time so the conduction current cannot establish a reversal, the charges trapped in the dielectric by the low mobility create an electric field by proximity with the foil edge that exceeds breakdown and partial discharges occur in the opposite direction, dissipating the excess space charge.

It is noted that because space charge formation due to charge injection is limited temporally by the magnitude of the free carrier mobility in the dielectric, a theoretically ideal impregnant would possess the electrical properties of high dielectric strength, low intrinsic/extrinsic carrier concentrations, and a high value for free carrier mobility so space charge formation times would be comparable or faster than the transient voltages applied to the foil edges.

Detection of High Frequency Partial Discharges

The HFD2, High Frequency Discharge Detection System developed at the Dielectrics Laboratory at SUNY Buffalo made possible the detection of PDs at max operating conditions of 2 kVpp steady state sinusoidal voltage, 100kHz frequency, 2000pF capacitance load and a pulse separation of less than 1us with a sensitivity of 1 kV or better. The schematic of the apparatus is shown in figure 6.

Effects of Thermal Aging on the DC Lifetimes of Polypropylene

Other work at the Dielectrics laboratory investigated the effects of thermal prestressig on the DC lifetimes and film

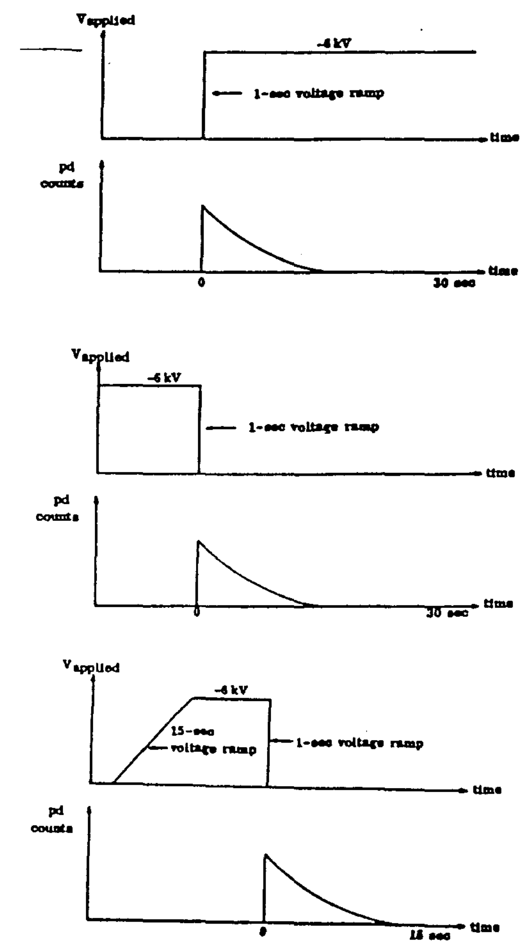


Figure 5. Partial Discharges at foil-edge structure vs. time as a function of applied voltage.

Attachment #1 - Page 3 of 5

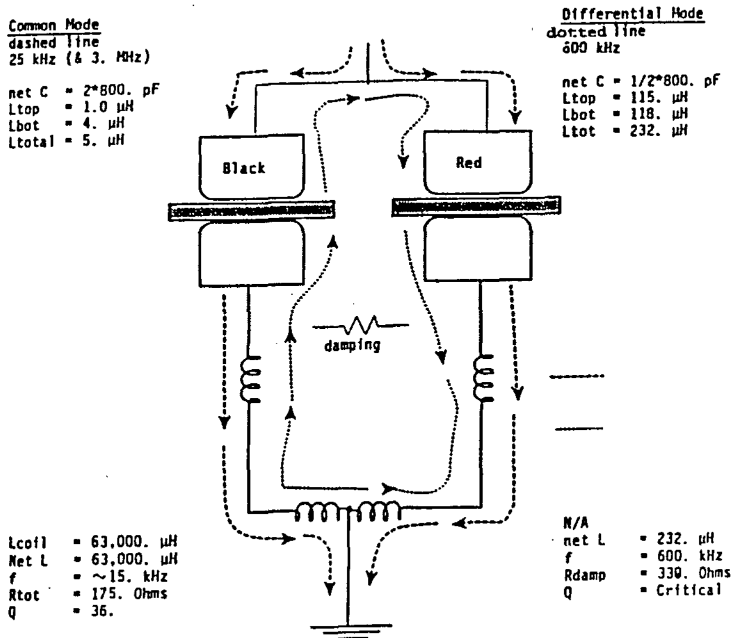


Figure 6. Schematic of torroidal bridge test fixture. Long dashes represent the current flow caused by the applied test voltage. This current should divide, with equal amounts going through each branch. The solid line represents the current path generated by discharge event. Note that the mutual coupling associated with each torroid opposes for the common mode current and adds for discharge current. The values shown represent measured values of the components for both common mode and differential model arrangements.

properties of polypropylene. Changes in surface properties (of the thermally aged vs. the control samples) were more pronounced that bulk effects. Methods for analyzing surface properties included Atomic Force Microscopy (AFM), Scanning Electron Microscopy (SEM) and Electron Scattering for Chemical Analysis (ESCA). The statistical analysis and fitting (to semi-empirical or quasi-theoretical life models) of the obtained replicate DC lifetime data could only

account for an 82% fit, i.e., input stress variables of applied voltage (DC field) and aging temperature level and duration can explain 82% of the variability in the data. The remaining 18% due to error and unknown factors that remain to be explained. Spatial variability at electrode-insulator interface and possible subsequent effects of heterocharge are a major factor in the obtained lifetimes.

Electromechanical Factors in Pulse Power Applications

In the case of high power pulse generators, for such high energy pulses, mechanical shock stress (during the puls or at its leading edge) seems to be the critical factor in the component survival. It was noted in [7] that as powe densities become very high, this shock may lead to anomalous behavior as well as introduce failure modes that ma not be immediately obvious. In general, the mechanical forces in the laminate components are generated by tw possible mechanisms; thermal shock and electrostatic and electromagnetic mechanisms.

The thermal shock (due to the fast deposition of thermal energy) leads to permanent restructuring of the insulators and sputter damage to metal surfaces both of which affect interfacial behavior and space charge characteristics. The temperature rise can be described by the three dimensional heat conduction equation:

$$\frac{\partial T}{\partial t} = D\nabla^2 T + \frac{D}{K}U \qquad (2)$$

where T is the temperature in Kelvin, t is the time in seconds, U is the power density on watts per cubic meter, D is a constant of diffusivity and K is the specific thermal conductivity of the material (in watts per second-meter-kelvin). The reduction of the equation to a one-dimensional form and solution under certain conditions given arious assumptions can be found in [7].

Figure 7 illustrates the transient temperature rise per unit input power in a dielectric film surrounded by a high diffusivity substrate as a function of pulse duration. Electrostatic forces also exert mechanical forces on the laminate components of a dielectric which may be expressed as

$$F = \frac{1}{2} V^2 \frac{dC}{dx} \qquad (3)$$

where V is the applied voltage, x is a dimension and C is the capacitance as a function of x. This mechanical force exerted by the metallic plates on the dielectric in a squeezing action that is transmitted to the oil (impregnant) and a pumping action is initiated during charging and discharging of the laminate. Thus the structure vibrates at the pulse repetition frequency with the force being predominant at localized areas of impurities. In the case the highest dielectric material does not fill complete volume between the conductors, a stretching force is exerted on the higher dielectric material in directions to fill the laminate volume. This means that any high dielectric impurities will be attracted to points/regions of highest field stress. For instance, oxides of aluminum do not have breakdown strength, but would be attracted to the highest fields leading to corona and eventually breakdown. From this it can be seen that although electrostatic forces are usually weak, nonetheless, they can lead to troublesome problems especially as energy densities increase as early and anomalous failure modes are observed.

Conclusions

The effects of space charge on prebreakdown and breakdown mechanisms are far from simple and need further investigation and modelling via a collaboration of theoretical and practical considerations and analysis. The impact on insulation aging would be very significant.

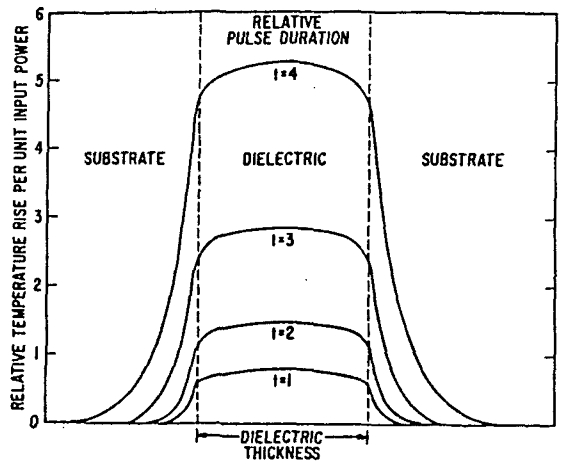


Figure 7. Illustration of the transient temperature rise per unit input in a dielectric film surrounded on either side by a thermally high diffusivity substrate. As the pulse duration increases, the thermal effects change from adiabatic heating to significant heat flow during the pulse.

References:

1] L. A. Dissado and J. C. Fothergill, Electrical Degradation and Breakdown in Polymers, Peter Peregrinus Ltd. London, 1992.

2] M. Ieda, "Carrier Injection, Space Charge and Electrical Breakdown in Insulating Polymers", IEEE Transactions on Electrical Insulation, Vol. 22, pp. 261-267, 1977.

3] T. Mizutani, "Space Charge Measurement Techniques and Space Charge in Polyethylene", IEEE Transactions on Dielectrics and Electrical Insulation, CEIDP, Vol. 1, No. 5, pp. 923-933, 1994.

4] R. W. Hare, R. M. Hill and C. J. Budd, "Modelling Charge Injection and Motion in Solid Dielectrics under High Electric Field", J. Physics. D: Applied Physics, Vol. 26, pp. 1084-1093, 1993.

5] M. Ieda, M. Nagato and M. Hikita, "High Field Conduction and Breakdown in Insulating Polymers. Present Situation and Future Prospects", IEEE Transactions on Dielectrics and Electrical Insulation, CEIDP, Vol. 1, No. 5, pp. 934-945, 1994.

6] W. J. Sarjeant, "Polymer Laminate Structures", Applied Physics Communications, 3(1&2), 1983.

7] T. R. Burkes and W. J. Sarjeant, "Electromechanical Shock in Pulse Power Components," Proc. Fourteenth Pulse Power Modulator Symposium, Orlando, Florida, June 3-5, pp. 125-129, 1980.

Supplément à la Revue "Le Vide : science, technique et applications" - N° 275, Mars 1995

RELATIONS BETWEEN ELECTRIC BREAKDOWN FIELD AND MECHANICAL PROPERTIES OF CERAMICS

S. CARABAJAR, C. OLAGNON, G. FANTOZZI, C. LE GRESSUS*
INSA - GEMPPM, URA 341, F69621 Villeurbanne Cedex.
* CEA-DAM, Centre de Bruyères le Châtel, F91680 France.

Relation between mechanical properties and electric breakdown is investigated. For this purpose, different ceramics, alumina with 2 grain sizes, zirconia, silicon nitride and mullite have been measured in terms of critical field and mechanical properties. The study has been conducted from a fracture mechanics point of view, i.e. rupture occurs from pre-existing flaws. The results show that different parameters play a role in the value of the critical field. The importance of the microstructure is demonstrated with the 2 aluminas that come from the same processing batch. Finally, the results are qualitatively described by a simple model based on the Griffith criteria.

Introduction:

The electric breakdown of ceramics has been extensively studied these last years on two different bases. On the one hand several theories have been developed to describe the different stages of breakdown on relatively model materials. On the orther hand different experiments have been conducted on samples or real components made of electric alumina or glass and other ceramic materials for electrical applications.

The electric breakdown is generally classified into two types: the surface breakdown and volumic breakdown. Different models based on different mechanisms have been proposed to describe the surface breakdown [1]. They however mostly present a commun series of steps as: (i) the initiation where the triple junction plays an important role, (ii) the development and (iii) the final discharge. The most significant differencies lying in the interpretation of the development step.

Different volumic breakdowns have also been observed[2,3]. They are generally divided into: (i) intrinsic breakdowns which is composed of electronic and electromechanical breakdown and (ii) thermal breakdown.

In both types of breakdowns or surface flashover different experimental parameters that are not always taken into account in models have been identified [4]. They are geometric, condition of applying the electric field (type, rate, discharge rate etc...), material (permitivity, surface roughness, homogeneity etc...), temperature, environemental dielectric.

The mechanical properties of polycristalline ceramic have also been extensively studied and described at high and room temperatures. It has been shown to be strongly dependant on the microstructure, i.e. the grain size, micro defects sizes (pores, cracks, grain boundaries) and impurities. The room mechanical properties of most polycristalline ceramics is mainly governed by their brittleness. The failure indeed occurs before any plastic deformation from the growth often unstable) of pre-existing microflaws. This results in a significant scattering in the distribution of strengths which must be used with a statistical treatment, mostly Weibull distribution.

AN INVESTIGATION OF AGING MECHANISMS AND STATISTICAL LIFE MODELS FOR POLYPROPYLENE FILMS

by

Amany N. Stokes

A Dissertation
Submitted to the
Faculty of the Graduate School
of the
State University of New York at Buffalo
in partial fulfillment of the requirements
for the degree
of
Doctor of Philosophy.

June 1995

ABSTRACT

An investigation is conducted to study and model the aging of polypropylene, a widely used polymer dielectric in energy storage and transport devices, under the sequential application of thermal and DC electrical stresses. Continuous thermal aging in air was carried out at temperatures of 90 °C and 110 °C for aging times of 100, 300, and 500 hours. The DC lifetimes of the control (thermally unaged) and thermally aged films were then obtained at four voltage stress levels of 16, 15, 14 and 13 kV.

Testing of a sufficiently large number of randomly chosen specimens in a well designed test plan allows for the analysis of all the effects of stress factors independently and interactively. A complete statistical analysis of the data included nonparametric and parametric assessment of adequate life distributions and life-stress models (linear and nonlinear) utilizing Least Squares and Maximum Likelihood regression methods. Several diagnostic tests, including Electron Scattering for Chemical Analysis (ESCA), Atomic Force Microscopy (AFM), Scanning Electron Microscopy (SEM), Fourier Transformed Infrared (FTIR) and Nuclear Magnetic Resonance (NMR) spectroscopy, Wide Angle X-ray Diffraction (WAXS), plus other mechanical and electrical characterizations including DC breakdown voltage, dielectric constant, dissipation factor, tensile stress and strain, and Young's Modulus, for the control and aged samples were carried out. Results were then compared to detect any correlating changes and interpreted in the light of available aging theories.

The obtained results revealed the possible contribution of thermal aging to morphological and mechanical variations in the film. Changes of surface properties of the thermally aged film samples were more pronounced than those of the bulk (compared to the control samples) which consequently lead to altered interfacial (electrode/insulator) characteristics. This combined with the inherent stochastic spatial and temporal inhomogeneities of the film samples and possible space charge effects under the constant DC field, resulted in the observation of localized interfacial breakdown (possibly electronic) for the thermally aged samples. On the other hand, a seemingly bulk limited breakdown of a mechanical nature was deduced for the control film samples. Statistically, thermal aging had a significant effect on the DC lifetimes of the film that contributed a *multiplicative effect interaction term* (between aging time and temperature) to the life-stress model.